

Adiabatic Condition for Nonlinear Systems

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We revisit the adiabatic criterion in stimulated Raman adiabatic passage for the three-level Λ -system, and compare the situation with and without nonlinearity. In linear systems, the adiabatic condition is derived with the help of the instantaneous eigenvalues and eigenstates of the Hamiltonian, a procedure that breaks down in the presence of nonlinearity. Using an explicit example relevant to photoassociation of atoms into diatomic molecules, we demonstrate that the proper way to derive the adiabatic condition for the nonlinear systems is through a linearization procedure.

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According to the adiabatic theorem of quantum mechanics [1], if the Hamiltonian is changed sufficiently slowly, then a system in a given non-degenerate eigenstate of the initial Hamiltonian (say, $|i\rangle$) evolves into the corresponding eigenstate of the instantaneous Hamiltonian without making any transitions. Here “sufficiently slowly” means that the rate of change of the Hamiltonian is much smaller compared to the level spacings:

$$|\dot{H}| \ll \hbar|\omega_{fi}|^2, \quad (\text{for any } f \neq i) \quad (1)$$

where ω_{fi} denotes the transition frequency between the instantaneous eigenstates $|i(t)\rangle$ and $|f(t)\rangle$. Condition (1) is referred to as the adiabatic condition.

The standard version of the adiabatic theorem, which has many applications in quantum state preparation and manipulation, applies to linear quantum systems. We, however, have noticed that the adiabatic condition (1) have been used in recent studies of nonlinear quantum gases. This may be problematic since the proof of the adiabatic condition makes explicit use of the concept of an orthonormal set of energy eigenstates and the linear superposition principle involving these states, both of which become invalid when nonlinearity is introduced into the system. The purpose of this paper is to provide a general method for deriving the adiabatic condition in nonlinear systems. This is achieved by considering a specific example of coherent population transfer in three-level quantum systems using the stimulated Raman adiabatic passage (STIRAP) method [2, 3].

Consider the three-level Λ -system schematically shown in Fig. 1. The excited state $|e\rangle$ is coupled to two ground states $|a\rangle$ and $|g\rangle$, with the coupling strengths (Rabi frequencies) denoted as Ω_p and Ω_d , respectively. Here the subscripts “ p ” (“ d ”) stand for “pump” (“dump”). In the interaction picture, under the two-photon resonance condition ($\delta = 0$), the Hamiltonian of this system reads

$$H_{\text{lin}} = -\hbar\Delta|e\rangle\langle e| + \frac{\hbar}{2}(\Omega_p|a\rangle\langle e| + \Omega_d|g\rangle\langle e| + h.c.) . \quad (2)$$

Without loss of generality, we will assume that the Rabi frequencies $\Omega_{p,d}$ are real and positive.

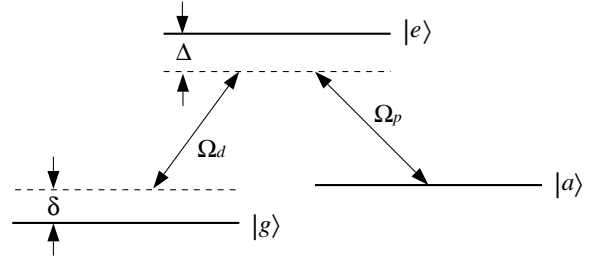


FIG. 1: The three-level system. Ω_p and Ω_d are the two coupling strengths, Δ and δ are one and two-photon detunings, respectively.

Hamiltonian (2) can be easily diagonalized. The particular energy eigenstate

$$|\text{CPT}\rangle = \frac{1}{\Omega_{\text{eff}}} (\Omega_d|a\rangle - \Omega_p|g\rangle) ,$$

where $\Omega_{\text{eff}} = \sqrt{\Omega_p^2 + \Omega_d^2}$, is known as the coherent population trapping (CPT) state or the dark state. The state $|\text{CPT}\rangle$ has zero eigenenergy and is decoupled from the excited state. When $\Delta = 0$, the other two eigenstates possess energies $\pm\hbar\Omega_{\text{eff}}/2$. When $\Omega_{p,d}$ are varied adiabatically, an initially prepared CPT state will remain in the instantaneous CPT state. A straightforward application of (1) leads to the adiabatic condition [3]

$$r_{\text{lin}} = \left| \frac{\dot{\Omega}_p\Omega_d - \dot{\Omega}_d\Omega_p}{\Omega_{\text{eff}}^3} \right| = \frac{|\dot{\chi}|}{1 + \chi^2} \frac{1}{\Omega_{\text{eff}}} \ll 1, \quad (3)$$

where $\chi \equiv \Omega_p/\Omega_d$. This property of the CPT state facilitates a coherent population transfer from $|a\rangle$ to $|g\rangle$ when χ changes from 0 to ∞ . This is the basis for STIRAP which has wide applications ranging from chemical reaction dynamics [4] to laser cooling of atoms [5].

In this paper, we want to derive the correct adiabatic condition for a nonlinear three-level system describing a

coupled atom-molecule system. This system has recently received significant attention due to the possibility of creating ultracold molecules by associating cold atoms [6, 7].

For the nonlinear system, we use the same level scheme of Fig. 1. Now state $|a\rangle$ represents an atomic state, while $|e\rangle$ and $|g\rangle$ are excited and ground diatomic molecular states. To keep things as simple as possible without sacrificing essential physics, we neglect nonlinear collisions between particles. The only nonlinearity comes from the fact that it takes two atoms to form a molecule. Under the two-photon resonance condition, the Hamiltonian in second quantized form reads

$$\hat{H}_{\text{nl}} = -\hbar\Delta\hat{\psi}_e^\dagger\hat{\psi}_e + \frac{\hbar}{2}\left(\Omega_p\hat{\psi}_e^\dagger\hat{\psi}_a\hat{\psi}_a + \Omega_d\hat{\psi}_e^\dagger\hat{\psi}_g + h.c.\right), \quad (4)$$

where $\hat{\psi}_i$ and $\hat{\psi}_i^\dagger$ are the annihilation and creation operators for state $|i\rangle$, respectively. In the mean-field treatment, these are replaced by c -numbers ψ_i and ψ_i^* , which obey the following coupled equations:

$$i\dot{\psi}_a = \Omega_p\psi_a^*\psi_e, \quad (5a)$$

$$i\dot{\psi}_e = \Delta\psi_e + \frac{\Omega_p}{2}\psi_a^2 + \frac{\Omega_d}{2}\psi_g, \quad (5b)$$

$$i\dot{\psi}_g = \frac{\Omega_d^*}{2}\psi_e, \quad (5c)$$

ψ_i 's are normalized as $|\psi_a|^2 + 2(|\psi_g|^2 + |\psi_e|^2) = 1$, a consequence of the conservation of total atom numbers.

To find the eigenstates of the system, we simply replace id/dt at the left hand sides of Eqs. (5) by ω , the eigenfrequency. It is easy to show, as in the linear counterpart, the nonlinear Λ -system supports a CPT eigenstate with zero eigenenergy [7], with the corresponding state vector given by $\Psi_0 = (\psi_a^0, \psi_e^0, \psi_g^0)^T$ where

$$\psi_a^0 = \left[\frac{2\Omega_d}{\Omega_d + \Omega_{\text{eff}}^{\text{nl}}}\right]^{1/2}, \quad \psi_e^0 = 0, \quad \psi_g^0 = -\frac{2\Omega_p}{\Omega_d + \Omega_{\text{eff}}^{\text{nl}}}. \quad (6)$$

with $\Omega_{\text{eff}}^{\text{nl}} \equiv \sqrt{\Omega_d^2 + 8\Omega_p^2}$. Evidence of the CPT state in coupled atom-molecule systems has been reported in several recent experiments [8].

Focusing on the case of vanishing single-photon detuning, $\Delta = 0$, we can easily obtain the other eigenstates:

$$\frac{1}{\sqrt{2}}(0, \pm 1, 1)^T,$$

with eigenfrequencies $\pm\Omega_d/2$, respectively. Furthermore, when $\Omega_d/\Omega_p < 1$, two more eigenstates appear:

$$\left(\sqrt{\frac{1}{2}\left(1 - \frac{\Omega_d^2}{\Omega_p^2}\right)}, \pm\frac{1}{2}, \frac{\Omega_d}{2\Omega_p}\right)^T,$$

with eigenfrequencies $\pm\Omega_p/2$, respectively. That there may exist more eigenstates than the dimension of the Hilbert space is unique for nonlinear systems [9]. The CPT state distinguishes itself from others with vanishing

probability amplitude in the excited state ψ_e . The non-orthogonality between the CPT state and any of the other eigenstates is quite obvious.

With the lack of a set of orthonormal energy eigenstates, the conventional linear adiabatic condition (1) or (3) can no longer be used for the nonlinear system. We want to derive the right adiabatic condition suitable for the nonlinear Λ -system under study. Our goal is to find the condition under which the system stays in the instantaneous CPT state (6) when the Rabi frequencies Ω_p and Ω_d are varied in time. To this end, we adopt the linear stability analysis which has wide application in various nonlinear systems.

To begin with, we expand the state vector as:

$$\psi_i = \psi_i^0 + \delta\psi_i,$$

where $\delta\psi_i$ represents the deviation of the probability amplitude in state $|i\rangle$ from the CPT solution (6). Adiabaticity is obeyed as long as these deviations remain small. Inserting this expansion into the dynamical Eqs. (5), we obtain the linearized equations:

$$i\frac{d}{dt}\delta\psi = \mathbf{M}\delta\psi + \dot{\Psi}_0, \quad (7)$$

where

$$\delta\psi = \begin{pmatrix} \delta\psi_a \\ \delta\psi_e \\ \delta\psi_g \end{pmatrix}, \quad \mathbf{M} = \begin{pmatrix} 0 & \Omega_p\psi_a^0 & 0 \\ \Omega_p\psi_a^0 & \Delta & \frac{\Omega_d}{2} \\ 0 & \frac{\Omega_d}{2} & 0 \end{pmatrix}.$$

The last term at the right hand side of (7) represents the ‘‘source’’ term arising from the temporal variation of Rabi frequencies ($\dot{\Omega}_p$ and $\dot{\Omega}_d$).

The eigenfrequencies of the linearized equations (i.e., the eigenvalues of matrix \mathbf{M}) can be easily found as

$$\omega_0 = 0, \\ \omega_{\pm} = \frac{1}{2} \left[\Delta \pm (\Delta^2 + \Omega_d \Omega_{\text{eff}}^{\text{nl}})^{1/2} \right].$$

The corresponding eigenstates, i.e., the normal modes, are

$$\mathbf{w}_0 = \mathcal{N}_0 \begin{pmatrix} -\Omega_d/2 \\ 0 \\ \Omega_p\psi_a^0 \end{pmatrix}, \quad \mathbf{w}_{\pm} = \mathcal{N}_{\pm} \begin{pmatrix} \Omega_p\psi_a^0 \\ \omega_{\pm} \\ \Omega_d/2 \end{pmatrix},$$

where $\mathcal{N}_{0,\pm}$ are normalization constants. Note that $\mathbf{w}_{0,\pm}$ are orthogonal to each other.

Since the frequencies $\omega_{0,\pm}$ are all real, in the limit of $\dot{\Omega}_{p,d} \rightarrow 0$ (i.e., the vanishing source terms), $\delta\psi_i$ will not grow in time and hence the system initially prepared in a CPT state will stay in the instantaneous CPT state. This is the adiabatic theorem generalized to this nonlinear system. We remark that for a general nonlinear system, complex eigenfrequencies of the linearized equations may appear [12]. These systems are *dynamically unstable* such that, even in the absence of source terms,

the deviations may grow from intrinsic quantum fluctuations [9, 10]. Adiabaticity will therefore break down in the dynamical unstable regimes of a nonlinear system.

The lack of complex eigenfrequencies in the current study guarantees that the system under consideration is always dynamically stable, hence the growth of deviations $\delta\psi_i$ can only be driven by the source terms.

From the first sight, the presence of the zero mode ω_0 seems to be most worrisome, since the fluctuation in this mode is energetically resonant with the CPT state. Fortunately, as we now show, this resonance does *not* lead to the growth of the zero mode. Writing the deviation vector as superpositions of normal modes

$$\delta\psi = \sum_{\alpha=0,\pm} c_\alpha \mathbf{w}_\alpha,$$

Equations (7) yield:

$$i\dot{c}_\alpha = \omega_\alpha c_\alpha - i \mathbf{w}_\alpha^\dagger \dot{\Psi}_0.$$

In particular, we have

$$i\dot{c}_0 = -i \mathbf{w}_0^\dagger \dot{\Psi}_0 = -i\mathcal{N}_0 \left(-\frac{\Omega_d}{2} \dot{\psi}_a^0 + \Omega_p \psi_a^0 \dot{\psi}_g^0 \right),$$

Using the CPT solution given in Eqs. (6), it is straightforward to show that the right hand side of the above equation vanishes and hence the amplitude of the zero mode fluctuation does not change in time. Therefore we reach a very important conclusion: Although the zero mode fluctuation is resonant with the CPT state, it is *not* coupled by the dynamics. From now on, we can simply ignore the zero mode and focus on the remaining two modes ω_\pm .

Assuming $c_\pm(0) = 0$, the amplitudes $c_\pm(t)$ can be solved as

$$c_\pm(t) = - \int_0^t dt' e^{i\omega_\pm(t'-t)} \mathbf{w}_\pm^\dagger(t') \dot{\Psi}_0(t'). \quad (8)$$

Defining the adiabaticity parameter as

$$r_{\text{nl}}(t) \equiv \frac{1}{2} \sqrt{|c_+(t)|^2 + |c_-(t)|^2}, \quad (9)$$

which represents the population in the fluctuations above the CPT solution, the adiabatic condition can now be defined quantitatively as

$$r_{\text{nl}}(t) \ll 1. \quad (10)$$

When ω_\pm is large, the exponential terms in (8) is rapidly oscillating and the most significant contribution of the integral comes from $t \approx t'$. We may change the time variable of $\mathbf{w}_\pm^\dagger(t') \dot{\Psi}_0(t')$ in the integrand to t and take it out of the integral, which yields

$$\begin{aligned} c_\pm(t) &= -\mathbf{w}_\pm^\dagger(t) \dot{\Psi}_0(t) \frac{1 - e^{-i\omega_\pm t}}{i\omega_\pm} \\ &= -\mathcal{N}_\pm \frac{\dot{\Omega}_p \Omega_d - \Omega_p \dot{\Omega}_d}{\Omega_d + \Omega_{\text{eff}}^{\text{nl}}} \frac{1 - e^{-i\omega_\pm t}}{i\omega_\pm}. \end{aligned}$$

The adiabatic condition now becomes

$$r_{\text{nl}}(t) \approx \frac{1}{2} \left(\frac{\mathcal{N}_+^2}{\omega_+^2} + \frac{\mathcal{N}_-^2}{\omega_-^2} \right)^{1/2} \frac{|\dot{\Omega}_p \Omega_d - \Omega_p \dot{\Omega}_d|}{\Omega_d + \Omega_{\text{eff}}^{\text{nl}}} \ll 1,$$

where we have neglected the oscillating term $e^{-i\omega_\pm t}$ in evaluating $r_{\text{nl}}(t)$.

For $\Delta = 0$, after some algebra, the above inequality leads to

$$r_{\text{nl}} = \frac{|\dot{\chi}|}{1 + \sqrt{1 + 8\chi^2}} \frac{1}{\Omega_{\text{eff}}^{\text{nl}}/2} \ll 1, \quad (11)$$

where $\chi = \Omega_p/\Omega_d$ as before. Comparing (11) with (3), we see that the adiabatic condition for the nonlinear system has a somewhat similar expression to that for the linear system, with, however, the following major difference: χ^2 in the denominator at the left hand side of (3) is replaced by $\sqrt{1 + 8\chi^2}$ in (11). It follows that, at the later stage of STIRAP where $\chi \gg 1$, the adiabatic condition for the nonlinear system is harder to fulfil than for its linear counterpart.

We remark that for the linear system, we can use the same linearization procedure to derive the adiabatic condition. It is obvious that, in this case, the eigenfrequencies of the linearized equations are the same as the ones of the original equations. Eqs. (9) and (10) will then reproduce the linear adiabatic condition (3).

To support the above analysis, we use the following numerical example. We choose two equal-amplitude equal-width Gaussian pulses for the pump and dump field centered at $t_{p,d}$, respectively,

$$\Omega_{p,d}(t) = \Omega_0 e^{-(t-t_{p,d})^2},$$

where we have taken the pulse width as the units for time. A counter-intuitive pulse sequence [11] requires that $t_d < t_p$. At the initial time ($t = 0$), only the $|a\rangle$ state is populated, i.e.,

$$\psi_a(0) = 1, \quad \psi_g(0) = \psi_m(0) = 0.$$

We evolve the state vector under the Hamiltonian H_{lin} and H_{ln} for the linear and nonlinear systems, respectively.

Figure 2 shows an example of the population dynamics. There are not much differences at the early stage for the linear and nonlinear cases. However, at the later stage of the STIRAP, the nonlinear system can no longer follow the CPT state while the linear system stays closely in the CPT state. In the end, we achieve an 80% population transfer in the former and near perfect transfer in the latter.

Figure 3 shows the corresponding time evolution of the adiabaticity parameter. At the early stage of STIRAP, the two parameters in the linear and nonlinear systems are almost identical. However, at the later stage, we have

$$r_{\text{nl}} \gg r_{\text{lin}},$$

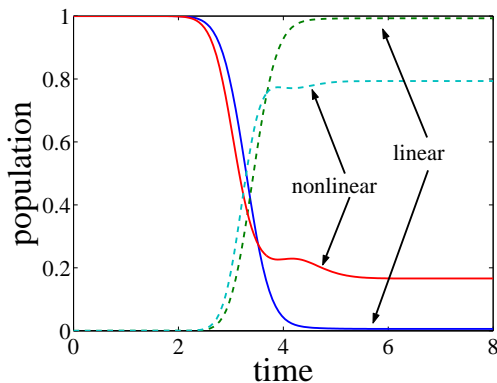


FIG. 2: (Color online) Population dynamics. Solid lines: population in state $|a\rangle$ ($|\psi_a|^2$); dashed lines: population in state $|g\rangle$ ($|\psi_g|^2$ for the linear case and $2|\psi_g|^2$ for the nonlinear case). The parameters used here are $\Delta = 0$, $\Omega_0 = 5$, $t_d = 3$ and $t_p = 3.8$.

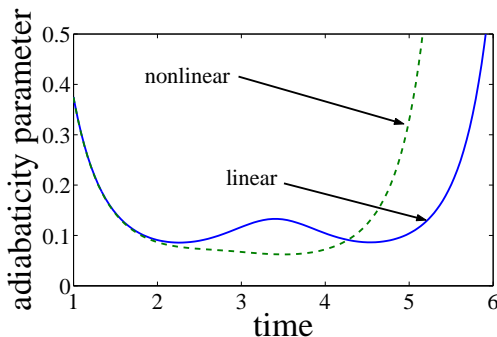


FIG. 3: (Color online) Adiabaticity parameter as defined in (3) and (11) for the linear and nonlinear systems, respectively. Same parameters as in Fig. 2.

in perfect agreement with our analysis, which also ex-

plains the reduced population transfer efficiency for the nonlinear system and the population dynamics as demonstrated in Fig. 2.

In conclusion, we have shown that, for nonlinear systems, the textbook method used in deriving the adiabatic condition fails to provide the correct answer, and the proper way to derive the adiabatic condition is through a linearization procedure. This is demonstrated by considering the coherent population transfer process via STIRAP in both a linear and a nonlinear three-level Λ -system, which are indeed governed by different adiabatic conditions. As a side, our analytical result of the nonlinear adiabatic condition (11) will become very useful in determining the required laser parameters for photoassociation via STIRAP.

As we have mentioned, in order to keep the mathematics as simple as possible, we have neglected here the nonlinear collisions between particles which are important in the ultracold quantum degenerate atomic/molecular systems. The inclusion of the collisions will introduce dynamically unstable regimes for the CPT states [12]. With collisions, the linearization procedure is equivalent to the Bogoliubov treatment of weakly interacting Bose condensates. However, the orthonormality of linearized eigenstates is replaced by *bi-orthonormality* of the Bogoliubov normal modes, and the zero-frequency mode corresponds to the Goldstone modes related to the spontaneously broken symmetries. These changes lead to considerably more complicated algebra. The adiabatic condition for collisional systems will be studied elsewhere.

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